Nonadiabatic transitions in a two-level quantum system: Pulse-shape dependence of the transition probability for a two-level atom driven by a pulsed radiation field

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The problem of a two-level atom interacting with a radiation pulse is studied in the limit that the atom-field detuning times the pulse duration is much greater than unity. Owing to the large atom-field detuning, transitions result from nonadiabatic coupling of the states by the field. The transition probability for the atom to be excited following the pulse is studied as a function of field strength for five different pulse shapes: hyperbolic secant, Lorentzian, hyperbolic secant squared, Lorentzian squared, and Gaussian. It is shown that the behavior of the transition probability differs *qualitatively* for these pulses. An explanation of this qualitative difference is given in terms of the Massey parameter. Numerical solutions are compared with asymptotic solutions and several anomalies are noted. In the limit of large field strength, a universal expression for the transition probability is found. An interesting feature of the solutions is that, in the limit of very large field strengths, the transition probability for a Gaussian pulse can approach unity despite the fact that the pulse has an exponentially small Fourier amplitude at the atom-field detuning. This apparent violation of the energy-time uncertainty principle is explained in terms of the nonlinear atom-field interactions. [S1050-2947(98)01801-0]

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I. INTRODUCTION

A basic problem in quantum mechanics is to determine the time evolution of a two-state quantum system whose dynamics is governed by an arbitrary, time-dependent Hermitian Hamiltonian. For example, one could imagine a spin-1/2 system in an external, pulsed magnetic field. The field results in a time-dependent coupling of the spin-up and spin-down states, as well as a time-dependent change in the energy separation of the levels. Although this problem appears to be deceptively simple, there exists only a very limited set of pulse shapes for which analytical solutions can be obtained for the state amplitudes. There is an extensive literature devoted to analytical and approximate methods for attacking this problem (often in the context of semiclassical approximations to a two-state scattering problem), some of which involve sophisticated mathematical techniques [1–7]. Of course, one can numerically solve the two, coupled differential equations for the state amplitudes, but, in certain limits, even such numerical methods pose serious challenges. It is our contention that this two-level problem still exhibits behavior that has yet to be fully understood and explored.

To prove our contention, we consider an even simpler problem, that of a two-level atom driven by a pulsed radiation field having an electric field vector of the form $\mathbf{E}=\mathbf{E}_0 f(t)\cos\omega t$. The field amplitude $\mathbf{E}_0$ and frequency ω are constant and the *smooth* field envelope function $f(t)$ has a temporal width of order T. It is assumed $f(t)$ and all its derivatives are continuous functions of t, $f(t)=f(-t)$, and that $f'(t)$ vanishes only at $t=0,\pm\infty$. It is furthermore assumed that $|\omega-\omega_0|/(\omega+\omega_0)\ll 1$, where ω_0 is the frequency separation between the levels, allowing one to make the rotating-wave or resonance approximation. Spontaneous decay during the pulse duration T is assumed to be negligible. With these assumptions, the atomic state amplitudes, written in an interaction representation, evolve as

$$\frac{da_1}{dt}=-i\beta f(t)\exp(i\alpha t)a_2, \tag{1a}$$

$$\frac{da_2}{dt}=-i\beta f(t)\exp(-i\alpha t)a_1. \tag{1b}$$

Here a_1 and a_2 represent the probability amplitudes associated with the atom's ground and excited levels, respectively, $\beta=-\mu E_0 T/2\hbar$ is a coupling strength (μ is a dipole moment matrix element), and $\alpha=(\omega-\omega_0)T$ is an atom-field detuning. All quantities, including the time, now expressed in units of T, are dimensionless. Without loss of generality, we assume that $\alpha\geqslant 0$ and $f(t)$ is normalized so that $\int_{-\infty}^{\infty} f(t)dt=1$. The quantity 2β is often referred to as the pulse area. Equations (1) define an effective two-level problem in which the coupling $\beta f(t)$ is time dependent, but the frequency separation of the levels α is constant.

For the given set of initial conditions

$$a_1(-\infty)=1, \tag{2a}$$

$$a_2(-\infty)=0, \tag{2b}$$

we are interested in determining the value of a_2 at $t=\infty$ and hence the behavior of the transition probability $P_2\equiv|a_2(\infty)|^2$ as we vary the pulse parameters $f(t)$ and β.

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Knowledge of the transition probability for different kinds of pulse shapes $f(t)$ and pulse strengths β is important, for example, when one needs to choose the appropriate pulse shape and pulse strength to maximize the transition probability for a given detuning. We are particularly interested in the so-called nonadiabatic limit

$$\alpha \gg 1. \tag{3}$$

When $\alpha \gg 1$, the field does not possess Fourier components to compensate effectively for the atom-field detuning. It is precisely the limit in which one might assert that the excitation probability is negligibly small following the pulse's action owing to the energy-time uncertainty principle.

The uncertainty principle argument certainly is correct in the perturbative limit $\beta \ll 1$, for which P_2 is exponentially small in the parameter α. However, as β increases in value, is the uncertainty principle argument still valid? One might expect that increasing the field *strength* does not compensate for the lack of Fourier components in the pulse at the atom-field detuning. This conclusion is reinforced by considering the transition probability for the Rosen-Zener [8] pulse envelope function $f(t)=1/2\ \mathrm{sech}(\pi t/2)$, the only smooth, symmetric pulse for which an analytic solution to Eqs. (1) has been found. The final-state amplitude and transition probability for this pulse envelope function are given by [9]

$$a_2(\infty)=-i\ \sin(\beta)\ \mathrm{sech}(\alpha), \quad P_2=\sin^2(\beta)\ \mathrm{sech}^2(\alpha). \tag{4}$$

Consistent with the uncertainty principle, the maximum transition probability is of order $\exp(-2\alpha)$, even for $\beta \gg 1$. Is this a general result, independent of pulse shape? The primary goal of this paper is to address this question. Using a dressed-atom approach, we show that an answer to this question can be given in terms of the Massey parameter [10] (to be defined below) associated with this problem. For certain pulse shapes the transition probability can be orders of magnitude greater than that predicted on the basis of the uncertainty principle. A secondary but equally important goal of this paper is to point out some mathematical anomalies that arise in asymptotic solutions of Eqs. (1).

In Sec. II we present solutions for the transition probability P_2, for different kinds of pulses, obtained by numerical means. In the limit of large α, one must take some care in numerically integrating Eqs. (1) since the integrands are rapidly varying on a (dimensionless) time scale of order unity. In Sec. III we give a qualitative explanation of the pulse shape dependence of the transition amplitudes in the limit of large coupling strength β. For $\beta \gg \alpha$, a universal form for the transition amplitude is given in terms of the Massey parameter. In Sec. IV we describe asymptotic methods that one can use to obtain P_2. In the limit of large α, we obtain approximate asymptotic solutions in the limits $\beta \ll \alpha$ and $\beta \gg \alpha$. Some mathematical anomalies associated with these asymptotic solutions are discussed. In Sec. V the results are summarized and the feasibility of experimental tests of the theory is explored. For those readers not interested in the mathematical details associated with the asymptotic solutions, Sec. IV can be omitted without loss of continuity.

II. NUMERICAL RESULTS

In this section we present solutions for P_2, for different kinds of pulses, obtained by numerical means. We concentrate on the following pulse shapes in this paper: hyperbolic-secant, $f_1(t)=(1/2)\ \mathrm{sech}(\pi t/2)$; Lorentzian, $f_2(t)=1/[\pi(1+t^2)]$; hyperbolic secant squared, $f_3(t)=(\pi/4)\ \mathrm{sech}^2(\pi t/2)$; Lorentzian squared, $f_4(t)=2/[\pi(1+t^2)^2]$; and Gaussian, $f_5(t)=(1/\sqrt{\pi})\exp(-t^2)$. In Fig. 1, the solid curves show how P_2 varies with β for these $f_i(t)$ $(i=1-5)$. We obtain P_2 by numerically integrating Eqs. (1), subject to the initial conditions (2).

Owing to the Rosen-Zener solution (4), one may be led to believe that any symmetric ''bell-shaped'' pulse, of which the hyperbolic secant is an example, will result in an atom acquiring a transition probability whose envelope is independent of β. However, we see from Fig. 1 that this is true only for the hyperbolic secant pulse and, at large pulse strengths, the hyperbolic secant squared pulse. For the Lorentzian and Lorentzian squared pulses, the transition probability decays to zero eventually, while for the Gaussian pulse, the envelope of the transition probability increases in the range of β studied [11]. For both the Lorentzian squared and hyperbolic secant squared pulses, the envelope of P_2 increases with increasing β for $r=2\beta/\alpha<1$, in agreement with the predictions of Robinson and Berman [12]. That the strong-field behavior of the transition probability should be so drastically different for these pulses, which are all rather similar in shape, is somewhat surprising. We seek to account for these differences in the next section.

III. PREDICTING THE TREND OF P_2 USING THE MASSEY PARAMETER

It is assumed that $\alpha \gg 1$. We find it convenient to work in the adiabatic or semiclassical dressed-state basis. To transform to the dressed-state basis, we first define new probability amplitudes $\tilde{a}_1$ and $\tilde{a}_2$ as

$$\tilde{a}_1(t)=a_1(t)\exp\left(-i\frac{\alpha}{2}t\right), \tag{5a}$$

$$\tilde{a}_2(t)=a_2(t)\exp\left(i\frac{\alpha}{2}t\right), \tag{5b}$$

from which we can then rewrite the two-state equations as

$$\frac{d\tilde{a}_1}{dt}=-i\frac{\alpha}{2}\tilde{a}_1-i\beta f(t)\tilde{a}_2, \tag{6a}$$

$$\frac{d\tilde{a}_2}{dt}=-i\beta f(t)\tilde{a}_1+i\frac{\alpha}{2}\tilde{a}_2. \tag{6b}$$

We then obtain the equations in the dressed-state representation by instantaneously diagonalizing the Hamiltonian

$$H_d=\begin{pmatrix} \alpha/2 & \beta f \\ \beta f & -\alpha/2 \end{pmatrix}. \tag{7}$$

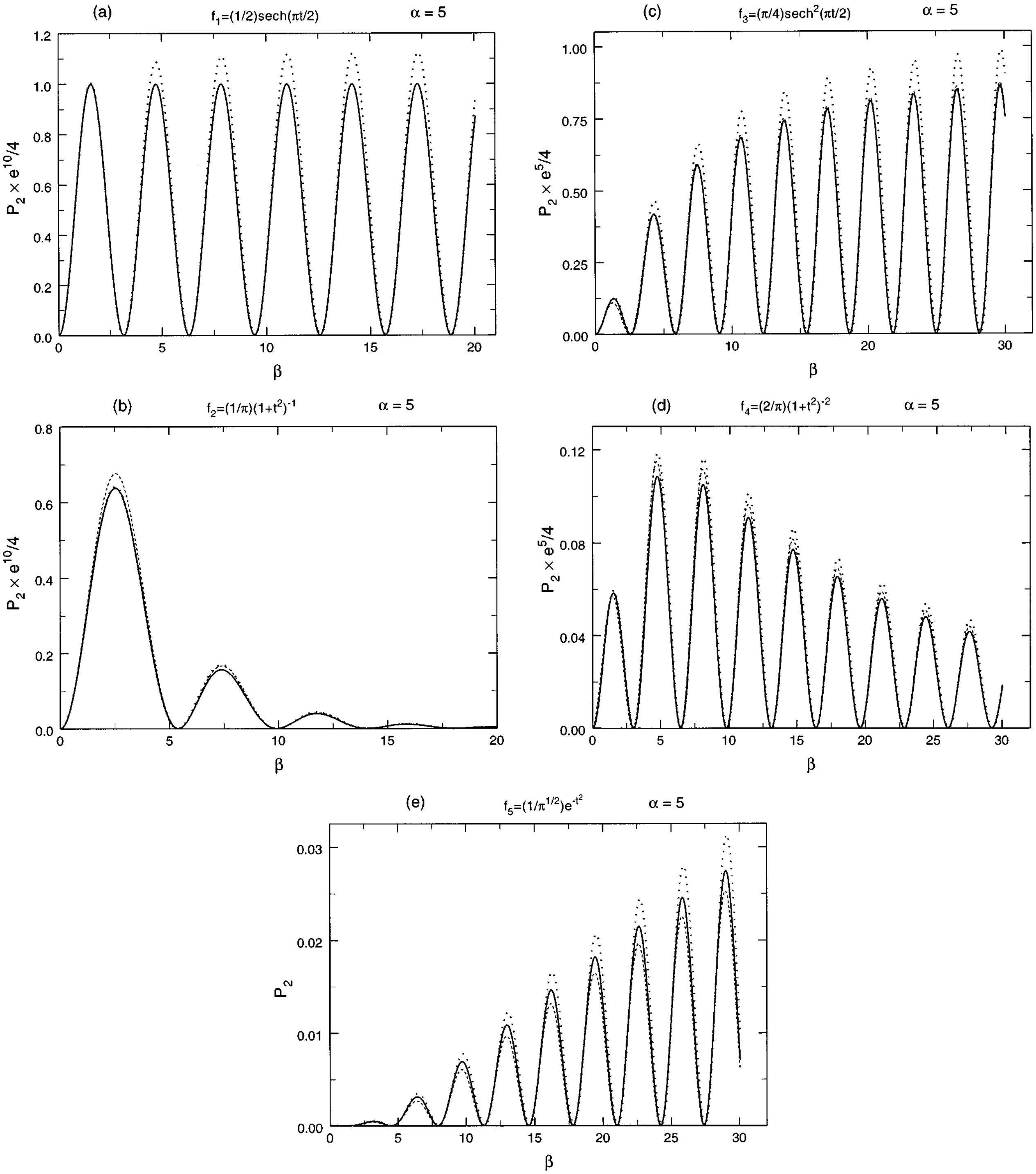

FIG. 1. Graph of the transition probability P_2 versus pulse strength β for a detuning $\alpha=5$. In each graph the exact numerical solution (solid line), asymptotic solution of the differential equations given by Eqs. (31) and (32) (dashed line), and perturbative solution in the dressed basis given by Eq. (27) (dotted line) are shown. The various pulse shapes considered are (a) hyperbolic secant, (b) hyperbolic secant squared, (c) Lorentzian, (d) Lorentzian squared, and (e) Gaussian. The hyperbolic secant and hyperbolic secant squared graphs are scaled to the asymptotic value for the envelope of P_2 predicted by Eqs. (38a) and (38c).

If we call $\tilde{b}_1$ and $\tilde{b}_2$ the probability amplitudes in the dressed basis, then the transformation to the dressed basis can be expressed through

$$\tilde{b}_1=\cos(\theta)\tilde{a}_1+\sin(\theta)\tilde{a}_2, \tag{8a}$$

$$\tilde{b}_2=-\sin(\theta)\tilde{a}_1+\cos(\theta)\tilde{a}_2, \tag{8b}$$

where the angle $\theta(t)$ is defined by

$$\sin[2\theta(t)]=\frac{2\beta f(t)}{\Omega(t)}, \tag{9}$$

$$\Omega(t)=\sqrt{\alpha^2+4\beta^2 f^2(t)}, \tag{10}$$

with $0 \leqslant \theta \leqslant \pi/4$. The time evolution of the dressed-state amplitudes is governed by the equations

$$\frac{d\tilde{b}_1}{dt} = -i\frac{\Omega(t)}{2}\tilde{b}_1 + \dot{\theta}\tilde{b}_2, \tag{11a}$$

$$\frac{d\tilde{b}_2}{dt} = -\dot{\theta}\tilde{b}_1 + i\frac{\Omega(t)}{2}\tilde{b}_2, \tag{11b}$$

which can be solved subject to the initial conditions $\tilde{b}_1(-\infty)=1$ and $\tilde{b}_2(-\infty)=0$ [9]. We note that Eqs. (9)–(11) are completely equivalent to Eqs. (1) and that

$$P_2 \equiv |a_2(\infty)|^2 = |\tilde{a}_2(\infty)|^2 = |\tilde{b}_2(\infty)|^2. \tag{12}$$

In fact, our numerical solutions are actually based on a solution of Eqs. (11) rather than Eqs. (1).

Equations (11) can be given a simple physical interpretation. The quantity $\Omega(t)$ is the instantaneous frequency separation of the dressed states. For $\alpha \neq 0$ the system starts in dressed state 1 at $t=-\infty$, when the frequency separation of the dressed states is α. As time evolves, the frequency separation of the levels increases, reaches a maximum at $t=0$, and then decreases, again achieving a value of α at $t=\infty$. The coupling between the dressed states is determined by the parameter

$$\dot{\theta} = \frac{r/2}{1+r^2 f^2}\frac{df}{dt}, \tag{13}$$

where

$$r = 2\beta/\alpha. \tag{14}$$

For $\alpha \gg 1$, the coupling parameter $\dot{\theta}$ does not possess Fourier components to compensate for the detuning $\Omega(t)$. As such, all transitions from dressed state 1 to 2 result from *nonadiabatic* coupling of the states. With increasing coupling strength, both the separation of the dressed states and the coupling between the dressed states increases. Whether or not the transition probability increases with increasing coupling strength depends on the relative increases of the coupling strength and level separation.

A measure of the nonadiabatic coupling of the states is provided by the Massey parameter defined by [10,1]

$$M(t) = \left|\frac{\Omega(t)}{\dot{\theta}}\right| = \left|\frac{\alpha[1+r^2 f^2(t)]^{3/2}}{r/2}\left(\frac{df}{dt}\right)^{-1}\right|. \tag{15}$$

For the problem under consideration in which $\alpha \gg 1$, one finds that the Massey parameter is much greater than unity, indicating that one is in the regime of nonadiabatic coupling [13]. It is not the magnitude of M that is of primary concern here. Rather it is the dependence of M on r and $f(t)$ that may provide us with some insight into the dependence of the envelope of P_2 as a function of r for a given f and α. For example, if $r \ll 1$, $M \sim |(2\alpha/r)(df/dt)^{-1}| \gtrsim 2\alpha/r$; regardless of the pulse shape, the nonadiabatic coupling increases with increasing r (that is, M decreases). This result is to be expected from a perturbative solution to Eqs. (11), where one finds that $\tilde{a}_2(\infty) \propto r$.

We are interested in explaining the qualitative differences that occur for different pulse shapes when $\alpha \gg 1$ and $r \gg 1$ $(\beta \gg \alpha)$. For $r \gg 1$, the minimum value of the Massey parameter occurs when $rf(t) \sim 1$. Thus it is reasonable to expect that a measure of the nonadiabatic coupling is given by the parameter

$$C = -\frac{r}{\alpha}\frac{df}{dt_0}, \tag{16}$$

where t_0 is the positive solution of

$$rf(t_0) = 1. \tag{17}$$

As r increases for a given α, one would expect an increasing envelope for P_2 if C increases, a decreasing envelope if C decreases, and a constant envelope if C is constant. Specifically, for the pulse shapes of Fig. 1, one finds the following.

$f_1(t) = (1/2)\ \mathrm{sech}(\pi t/2)$: $C \sim 1/\alpha$. The frequency separation of the two dressed states grows at the same rate as does the coupling strength when β is increased. As a result of this lack of dependence of C on r, one expects the transition probability to saturate at a constant value with increasing β. This conclusion, of course, is consistent with Eq. (4) and Fig. 1(a).

$f_2(t) = 1/[\pi(1+t^2)]$: $C \sim 1/(\alpha\sqrt{r})$, which decreases with increasing r. In this case, the frequency separation of the two adiabatic states grows faster than the coupling strength as β is increased. Thus, at large β, the nonadiabatic coupling is expected to decrease. Referring to Fig. 1(b), we see that this is true: the transition probability decays as β increases.

$f_3(t) = (\pi/4)\ \mathrm{sech}^2(\pi t/2)$: $C \sim 1/\alpha$, which is independent of β. By a similar argument as that given for the hyperbolic-secant pulse, we expect that the transition probability should saturate to a constant value. This agrees with the results shown in Fig. 1(c).

$f_4(t) = 2/[\pi(1+t^2)^2]$: $C \sim 1/\alpha r^{1/4}$, which decreases as r increases. Employing a similar argument as that for the Lorentzian pulse, we see that the transition probability should decay to zero also for large β. This is what is observed in Fig. 1(d). In addition, we see that C falls off more slowly with increasing r than in the case of the Lorentzian pulse ($r^{-1/4}$ as compared to $r^{-1/2}$). This weaker dependence of C on r translates to a weaker decrease in the transition probability as β is increased. Comparing the results in Figs. 1(c) and 1(d), we see that the decay in P_2 is indeed more gradual for the Lorentzian-squared pulse than it is for the Lorentzian pulse.

$f_5(t) = 1/\sqrt{\pi}\exp(-t^2)$: $C \sim \sqrt{\ln r}/\alpha$, which increases as r increases for $r > 1$. In this case, the nonadiabatic coupling increases at a faster rate than the frequency separation of the levels. This means that the transition probability should increase as β is increased, in agreement with the results shown in Fig. 1(e) [11,13].

Thus, using this approach, by investigating the dependence of the Massey parameter M on the pulse strength β, we are able to qualitatively account for the strong-field behavior of the transition probability as a function of pulse shape. In Sec. IV we obtain asymptotic expressions for P_2 in the limits $\alpha \gg 1$ and $r \gg 1$, which are in agreement with these qualitative findings. Moreover, we show that there appears to be a universal form for the transition amplitude in this limit, which can be expressed as

$$a_2(\infty)\sim-2ie^{-K\mathrm{Im}\mathfrak{M}(z_c^0)}\sin[\beta-K\mathrm{Re}\mathfrak{M}(z_c^0)], \quad (18)$$

where

$$\mathfrak{M}(z_c^0)\sim-\alpha/(r\ df/dz_c^0) \quad (19)$$

is proportional to the minimum value of the Massey parameter when $r\gg1$, z_c^0 is the solution of $\Omega(z)=\sqrt{\alpha^2+4\beta^2f^2(z)}=0$ that lies in the first quadrant of the complex-z plane and is closest to the real axis, and K is a constant of order unity that varies with pulse shape. With Eqs. (18) and (19), one achieves an amazingly compact form for the dependence of the transition amplitude in the limits of large detuning and large coupling strength $\alpha,r\gg1$. This was the primary goal of this work.

IV. ASYMPTOTIC APPROACHES

In this section we examine a few approximate solutions to the equations for the probability amplitudes in the limit of large α. These approximation techniques are by no means exhaustive, but they reveal several interesting anomalies in the solutions that have yet to be resolved. The methods to be discussed are (A) perturbative solution in the normal basis, (B) first-order perturbative solution in the dressed basis [(1) series solution in β, (2) asymptotic solution for arbitrary β, and (3) numerical solution for arbitrary β], and (C) asymptotic solutions of the differential equations [(1) solution for $\beta\gg\alpha$, (2) solution for $\beta\ll\alpha$, and (3) numerical solution for arbitrary β].

A. Perturbative solution in the normal basis

It is possible to solve Eqs. (1) iteratively to obtain $a_2(\infty)$ as a series in odd powers of β. To lowest order in β, the solution is proportional to the Fourier transform of the pulse envelope function. It is not possible, in general, to obtain analytical solutions for terms of order β^3 or higher, but approximate expressions can be obtained in some cases when $\alpha\gg1$. A method for obtaining such solutions is given in the Appendix, in which the terms of order β^3 and β^5 have been calculated to lowest nonvanishing order in $1/\alpha$. For the hyperbolic secant pulse, one finds

$$a_2(\infty,hs)\sim-2ie^{-\alpha}(\beta-\beta^3/3!+\beta^5/5!-\cdots), \quad (20)$$

which coincides with the solution (4) in the limit $\alpha\gg1$. Note that this series, which is an approximate solution to the equations of motion in the limit $\alpha\gg1$, converges to the correct asymptotic solution for all powers of β. For the Lorentzian pulse, one obtains

$$a_2(\infty,lz)\sim-2ie^{-\alpha}\left[\frac{\beta}{2}-\left(\frac{1}{3!}\right)\left(\frac{\beta}{2}\right)^3+\left(\frac{1}{5!}\right)\left(\frac{\beta}{2}\right)^5-\cdots\right]. \quad (21)$$

The two results differ by only by a scale factor in β, as predicted by Robinson and Berman [12] for pulses whose Fourier transform is of the form $\exp(-\alpha)$ for large α. Note, however, that Eq. (21) is *not* asymptotically correct for all β since it does not exhibit the falloff with increasing β shown in Fig. 1(b); the approximate solution (21) is valid only for $\alpha\gg1$, $r<1$, and $r\beta=(2\beta/\alpha)\beta\ll1$ [14]. In the case of the hyperbolic secant squared pulse, we find

$$a_2(\infty,hs^2)\sim-2i\pi e^{-\alpha}\left[\left(\frac{\alpha\beta}{\pi}\right)-\left(\frac{10-\pi^2}{6}\right)\left(\frac{\alpha\beta}{\pi}\right)^3+3.505\times10^{-5}\left(\frac{\alpha\beta}{\pi}\right)^5-\cdots\right], \quad (22)$$

valid for $\alpha\gg1$ and $r\sqrt{\alpha\beta}\ll1$ [14], and, for the Lorentzian squared pulse,

$$a_2(\infty,lz^2)\sim-2i\pi e^{-\alpha}\left[\left(1+\frac{1}{\alpha}\right)\left(\frac{\alpha\beta}{2\pi}\right)-\left(\frac{10-\pi^2}{6}\right)\left(\frac{\alpha\beta}{2\pi}\right)^3+3.505\times10^{-5}\left(\frac{\alpha\beta}{2\pi}\right)^5-\cdots\right], \quad (23)$$

valid for $\alpha\gg1$, $r<1$, $r\beta\ll1$, and $r\sqrt{\alpha\beta}\ll1$ [14]. The asymptotic forms, which again differ only by a scale factor [12] for $\alpha\gg1$, can no longer be factored into separate functions of α and β. As is to be seen below, these terms represent an envelope function for the transition probability that increases with increasing β for $r<1$. The calculations for the Gaussian pulse are somewhat more complicated since the Gaussian does not have a simple pole structure. Nevertheless, it is still possible to carry out an iterative solution to obtain

$$a_2(\infty,gs)\sim-i\left[\beta e^{-\alpha^2/4}-\left(\frac{9\beta^3e^{-\alpha^2/12}}{4\sqrt{3}\pi\alpha^2}\right)+\left(\frac{625\beta^5e^{-\alpha^2/20}}{64\sqrt{5}\pi^2\alpha^4}\right)-\cdots\right], \quad (24)$$

valid for $\alpha\gg1$ and $\beta\ll\alpha e^{\alpha^2/140}$ [see Eq. (A16e) of the Appendix]. Note that for large α, the third-order contribution can become larger than the first-order one even for values of β less than unity. The origin of this effect can be traced to the fact that the Fourier spectrum of the third-order coupling is significantly broader than that of the linear coupling term.

B. First-order perturbative solution in the dressed basis

By defining b_1 and b_2 using

$$\tilde{b}_1(t)=b_1\exp\left(-i\int_0^t\frac{\Omega(t')}{2}dt'\right), \quad (25a)$$

$$\tilde{b}_2(t)=b_2\exp\left(i\int_0^t\frac{\Omega(t')}{2}dt'\right), \quad (25b)$$

one can transform Eqs. (11) into the form

$$\frac{db_1}{dt}=\dot{\theta}\exp\left(i\int_0^t\frac{\Omega(t')}{2}dt'\right)b_2, \quad (26a)$$

$$\frac{db_2}{dt} = -\dot{\theta}\exp\left(-i\int_0^t \frac{\Omega(t')}{2}dt'\right)b_1. \qquad (26b)$$

From Eqs. (2), (5), and (8), it follows that the initial conditions are $b_1(-\infty)=e^{-i\Phi}$ and $b_2(-\infty)=0$, where $\Phi = \int_0^\infty [\Omega(t)-\alpha]dt$ [9]. Since the dressed states account for the rapid phase oscillations of the atom-field interaction and since the entire coupling to dressed state 2 has as its origin nonadiabatic transitions from state 1 to state 2, it is not unreasonable to believe that an accurate solution to our problem can be obtained by solving these equations *to first order* in the coupling constant $\dot{\theta}$, namely

$$\begin{aligned} b_2^{(1)}(\infty;ad) &= e^{-i\Phi}a_2^{(1)}(\infty;ad) \\ &= -e^{-i\Phi}\int_{-\infty}^{+\infty}\dot{\theta}\exp\left(-i\int_0^t \Omega(t')dt'\right)dt \\ &= -e^{-i\Phi}\int_{-\infty}^{+\infty}dt\,\frac{r/2}{1+r^2f^2}\,\frac{df}{dt} \\ &\quad\times\exp\left(-i\alpha\int_0^t\sqrt{1+r^2f^2(t')}dt'\right) \\ &= -e^{-i\Phi}\int_{-\infty}^{+\infty}dt\,\frac{\alpha\beta}{\alpha^2+4\beta^2f^2}\,\frac{df}{dt} \\ &\quad\times\exp\left(-i\int_0^t\sqrt{\alpha^2+4\beta^2f^2(t')}dt'\right). \end{aligned} \qquad (27)$$

It is important to note that a solution to first-order in $\dot{\theta}$ is a solution to *all orders* in the coupling strength β. One would expect corrections to first-order perturbation theory to be of order $1/\alpha$ since $\dot{\theta}\sim 1/\alpha$ for $\alpha \gg 1$, but, as we shall see, this is not necessarily the case.

1. Series solution in β

The integrand in Eq. (27) can be expanded as a power series in β and then integrated term by term. Details are given in the Appendix. For $\alpha \gg 1$ and $r=2\beta/\alpha \ll 1$, one obtains

$$\begin{aligned} a_2^{(1)}(\infty,hs;ad) \sim -2ie^{-\alpha}[&\beta - A(hs;3)\beta^3/3! \\ &+A(hs;5)\beta^5/5! - \cdots], \end{aligned} \qquad (28a)$$

$$\begin{aligned} a_2^{(1)}(\infty,lz;ad) \sim -2ie^{-\alpha}&\left[\frac{\beta}{2} - A(lz;3)\left(\frac{1}{3!}\right)\left(\frac{\beta}{2}\right)^3 + A(lz;5)\right. \\ &\left.\times\left(\frac{1}{5!}\right)\left(\frac{\beta}{2}\right)^5 - \cdots\right], \end{aligned} \qquad (28b)$$

$$\begin{aligned} a_2^{(1)}(\infty,hs^2;ad) \sim -2i\pi e^{-\alpha}&\left\{\left(\frac{\alpha\beta}{\pi}\right) - A(hs^2;3)\right. \\ &\times\left[\left(\frac{10-\pi^2}{6}\right)\left(\frac{\alpha\beta}{\pi}\right)^3\right] + A(hs^2;5) \\ &\left.\times\left[3.505\times 10^{-5}\left(\frac{\alpha\beta}{\pi}\right)^5\right] - \cdots\right\}, \end{aligned} \qquad (28c)$$

$$\begin{aligned} a_2^{(1)}(\infty,lz^2;ad) \sim -2i\pi e^{-\alpha}&\left\{\left(1+\frac{1}{\alpha}\right)\left(\frac{\alpha\beta}{2\pi}\right) - A(lz^2;3)\right. \\ &\times\left[\left(\frac{10-\pi^2}{6}\right)\left(\frac{\alpha\beta}{2\pi}\right)^3\right] + A(lz^2;5) \\ &\left.\times\left[3.505\times 10^{-5}\left(\frac{\alpha\beta}{2\pi}\right)^5\right] - \cdots\right\}, \end{aligned} \qquad (28d)$$

$$\begin{aligned} a_2^{(1)}(\infty,gs;ad) \sim -i&\left[\beta e^{-\alpha^2/4} - A(gs;3)\left(\frac{9\beta^3 e^{-\alpha^2/12}}{4\sqrt{3}\pi\alpha^2}\right)\right. \\ &\left.+A(gs;5)\left(\frac{625\beta^5 e^{-\alpha^2/20}}{64\sqrt{5}\pi^2\alpha^4}\right) - \cdots\right], \end{aligned} \qquad (28e)$$

where

$$A(hs;3) = A(lz;3) = 10/\pi^2 \approx 1.01, \qquad (29a)$$

$$A(hs;5) = A(lz;5) = \left(\frac{298}{300}\right)\left(\frac{10}{\pi^2}\right)^2 \approx 1.02, \qquad (29b)$$

$$A(hs^2;3) = A(lz^2;3) = \left(\frac{2}{15}\right)\left(\frac{1}{10-\pi^2}\right) \approx 1.02, \qquad (29c)$$

$$A(hs^2;5) = A(lz^2;5) = \left(\frac{2^4}{9!}\right)\left(\frac{516}{630}\right)\left(\frac{1}{3.505\times 10^{-5}}\right) \approx 1.03, \qquad (29d)$$

$$A(gs;3) = \left(\frac{28}{27}\right) \approx 1.04, \qquad (29e)$$

$$A(gs;5) = \frac{51}{5}\,\frac{64}{625} \approx 1.04. \qquad (29f)$$

(In the Appendix, the terms of order β^3 are calculated for arbitrary α.) If the A's were equal to unity, Eqs. (28) would coincide with Eqs. (20)–(24). The fact that all the A's are nearly equal to unity indicates that *first-order* perturbation theory in the dressed basis reproduces with high accuracy term by term the iterative solution (20)–(24) of the exact equations (1).

There are some anomalies in the solution, however. Note that the A's are independent of α, indicating that the term-by-term corrections do not decrease with increasing α as had been anticipated. One can show that by carrying out an *iterative* solution of Eqs. (26) in powers of $\dot{\theta}$ each successive term in the iteration corrects the corresponding term in the series solutions (28). For example, by going to order $\dot{\theta}^3$ (only odd orders enter the solution for a_2), one finds that all the coefficients of the β^3 terms agree *exactly* with the corresponding terms of the iterative solutions (20)–(24); by going to fifth order in $\dot{\theta}$, the coefficients of the β^5 terms agree, etc. Although the coefficients of the individual terms in Eqs. (28) do not agree exactly with the corresponding terms of the iterative solution, it is still possible that first-order perturbation theory in the dressed basis can provide a good approximation to the *exact* solution of Eqs. (1). We now turn our attention to this question.

2. Asymptotic solution for arbitrary β

It is possible to evaluate the integral (27) by the method of steepest descents. Since the techniques involved are similar to those encountered in asymptotic solutions of the differential equations (26) that are discussed below, we defer a detailed discussion of the steepest-descent method for the time being. It is interesting, however, to give the result of the steepest-descent calculation for the hyperbolic secant pulse. In that case, one finds [15]

$$a_2^{(1)}(\infty,hs;ad) \sim -2i\left(\frac{\pi}{3}\right)e^{-\alpha}\sin\beta. \qquad (30)$$

Aside from the overall factor of $\pi/3$, this result agrees with the *exact* asymptotic result (4). That the prefactor is not given correctly has been pointed out by Nikitin and Umanskii [1] and Davis and Pechukas [4], among others. Nikitin and Umanskii state that this is related to the fact that the integrand is not an analytic function of the Massey parameter. The true prefactor can be obtained by comparison with the solution of an exactly solvable problem. Putting aside any problems related to the prefactor, the solution (30) presents an interesting mathematical anomaly. The $\sin\beta$ dependence in Eq. (30) agrees with the exact asymptotic result (4), but *not* with the series expansion (28a) of the *integral* (27), which has been evaluated by the steepest-descent method to arrive at the result (30). In other words, it does not appear that the standard steepest-descent evaluation of the integral (27) can reproduce the series (28a). As such, one must consider the correct asymptotic evaluation of the integral (27) for $\alpha \gg 1$ and arbitrary β as an open problem.

3. Numerical solution for arbitrary β

The integral (27) can be evaluated numerically for the various pulse shapes considered in this paper. The numerical evaluation of the integral can be more time consuming than a direct solution of the differential equations (11), so there is little benefit to be gained by using the first-order result in the dressed basis if one is interested in exact results. Nevertheless, it is of some interest to compare the first-order perturbative results in the dressed basis with the exact results. The first-order perturbative results in the dressed basis are shown as the dotted lines in Fig. 1. As seen in the figures, the first-order perturbative solution in the dressed basis qualitatively tracks the exact solution, but overestimates the transition probability at large β. For $\beta \ll 1$, the integral solution (27) reproduces the perturbative results (20)–(24). For $\beta \gg 1$, the relative error in the solution appears to saturate in the range of β that we studied. Note that a *correct* asymptotic evaluation of the integral (27) must reproduce its almost perfect agreement with the exact solution for $\beta \ll 1$ and its deviations from the exact solution for $\beta \gtrsim 1$.

C. Asymptotic solutions of the differential equations

Crothers [2], Davis and Pechukas [4], and Nikitin and Umanskii [1] employ steepest-descent methods to solve Eqs. (26). For symmetric pulse shapes and $\alpha \gg 1$, the transition amplitude is given by [15]

$$a_2(\infty) \sim -2ie^{-y_a}\sin x_a, \qquad (31)$$

where

$$z_a \equiv x_a + iy_a = \int_0^{z_c^0} \sqrt{\alpha^2 + 4\beta^2 f^2(z)}\,dz \qquad (32)$$

and z_c^0 is the zero of the integrand lying closest to the real axis in the first quadrant of the complex-z plane. Crothers [2] presents a modified version of the result

$$a_2(\infty) \sim -i\ \mathrm{sech}\,y_a \sin x_a, \qquad (33)$$

which coincides with Eq. (31) when $y_a \gg 1$. The relative corrections to Eqs. (31) and (33) depend on the specific pulse shape, but decrease, at worst, as an inverse power of α. In general, the errors also decrease with increasing β. In order to understand the approximations that are used to obtain Eq. (31), one must examine the zeros z_c of $\alpha^2 + 4\beta^2 f^2(z)$. In the steepest-descent method the major contributions to the solution come from regions around the zeros in the upper half plane since these are the saddle points of the integral form of the differential equations. For the solution (31) to be valid, the zeros must be separated by a distance much greater than α^{-1}, so that the dominant contribution to the solution will be from the zeros closest to the real axis in the upper half plane. For symmetric pulses, if there is a zero at $x_c + iy_c$, there is also one at $-x_c + iy_c$. The contributions from these two zeros combine to give the sine function in Eq. (31), but, in principle, the zeros at $\pm x_c + iy_c$ must also be well separated for Eq. (31) to remain valid. For the pulse shapes $f_i(t)$ ($i = 1-5$) considered in this work, the zeros z_c and zeros closest to the real axis in the first quadrant z_c^0 are given by [16]

$$z_c(hs) = \pm i \pm (2/\pi)\ln\left[\frac{\beta}{\alpha} + \sqrt{1 + \left(\frac{\beta}{\alpha}\right)^2}\right],$$
$$z_c^0(hs) = i + (2/\pi)\ln\left[\frac{\beta}{\alpha} + \sqrt{1 + \left(\frac{\beta}{\alpha}\right)^2}\right], \qquad (34a)$$

$$z_c(lz) = \pm i\sqrt{1 \pm i\left(\frac{2\beta}{\pi\alpha}\right)}, \quad z_c^0(lz) = i\sqrt{1 - i\left(\frac{2\beta}{\pi\alpha}\right)}, \qquad (34b)$$

$$z_c(hs^2) = \pm(2/\pi)\ln(\pm\sqrt{ir_3} + \sqrt{ir_3 - 1}),$$
$$r_3 = \pm\left(\frac{\pi\beta}{2\alpha}\right),$$
$$z_c^0(hs^2) = (2/\pi)\ln\left[\sqrt{i\left(\frac{\pi\beta}{2\alpha}\right)} + \sqrt{i\left(\frac{\pi\beta}{2\alpha}\right) - 1}\right], \qquad (34c)$$

$$z_c(lz^2)=\pm i\left[1\pm e^{\pm i\pi/4}\sqrt{\left(\frac{4\beta}{\pi\alpha}\right)}\right]^{1/2},$$

$$z_c^0(lz^2)=i\left(1-e^{i\pi/4}\sqrt{\frac{4\beta}{\pi\alpha}}\right)^{1/2}, \quad (34d)$$

$$z_c(gs)=\pm\left[\ln\left(\frac{2\beta}{\sqrt{\pi}\alpha}\right)+(2n+1)i\frac{\pi}{2}\right]^{1/2} \quad \{n,-\infty,\infty\},$$

$$z_c^0(gs)=\left[\ln\left(\frac{2\beta}{\sqrt{\pi}\alpha}\right)+i\frac{\pi}{2}\right]^{1/2}. \quad (34e)$$

The hyperbolic secant and Lorentzian pulses have a single zero in the first quadrant [16], the hyperbolic secant squared and Lorentzian squared pulses have two zeros in the first quadrant [16], and the Gaussian pulse has an infinite number of zeros in the first quadrant.

It is easy to verify that, when $\alpha\gg 1$, the zeros in the lower half plane are well separated from those in the upper half plane, as is required for the validity of Eq. (31). For $\beta\gg\alpha$, the zeros in the upper half plane are well separated for each pulse and one would expect Eq. (31) to be valid. This conclusion is not strictly true for the Gaussian pulse; when $\pi/\sqrt{\ln(2\beta/\sqrt{\pi}\alpha)}\ll 1$, the infinite number of zeros in each quadrant coalesce and the zeros in the first quadrant coalesce with those in the fourth, as do those in the second and third quadrants. Nevertheless, owing to the logarithmic behavior, there is a wide range of β for which the zeros are well separated. On the other hand, it is also easy to verify that, for each pulse shape, *all* the zeros in the upper half plane coalesce for $\beta\ll\alpha$, implying that the result (31) cannot be relied upon in this perturbative limit. It is possible to modify the asymptotic procedure to allow for coalescing zeros (and for weighting functions $\dot\theta$ that possess poles at the position of the coalescing zeros) [3,17,18], but we have not carried out such calculations for this problem.

For the hyperbolic secant pulse, $z_c^0=\beta+i\alpha$ [2]. Generally speaking, however, Eq. (32) must be evaluated numerically. In contrast to the numerical evaluation of the integral (27) or the numerical solution of the differential equations (11), one does not encounter problems relating to an oscillating integrand in Eq. (32), allowing one to easily obtain values for z_a for arbitrarily large α and β. This is an important consideration if one wishes to obtain reasonably good values for the transition probability in the limits of very large α and β without extensive and difficult numerical computation. It is also possible to obtain expressions for z_a in the limit of large and small $r=2\beta/\alpha$. As noted above, the asymptotic forms in the limit of small r may have a limited range of validity owing to the fact that the various zeros z_c are not well separated in the complex plane. Nevertheless, results for this limit are given along with some suggestions for improving these results.

1. Asymptotic solution for $r\gg 1$

When $r\gg 1$, one can expand Eq. (32) as

$$z_a=\alpha\int_0^{z_c^0}dz\left(rf(z)+\sum_{n=1}^{\infty}\frac{(-1)^{n+1}(2n-1)!!}{2^n n!(2n-1)[rf(z)]^{2n-1}}\right)$$

$$=\beta-\alpha\int_{z_c^0}^{\infty}dz\; rf(z)$$

$$+\alpha\sum_{n=1}^{\infty}\frac{(-1)^{n+1}(2n-1)!!}{2^n n!(2n-1)}\int_0^{z_c^0}dz\frac{1}{[rf(z)]^{2n-1}}, \quad (35)$$

where the equalities $\int_0^\infty dz\, f(z)=1/2$ and $r=2\beta/\alpha$ have been used. Each term is then evaluated to give the leading contribution when $r\gg 1$. For a pulse shape that varies as $bt^{-\mu}$ $(\mu>1)$ for $|t|\gg 1$ [as do the Lorentzian $(b=1/\pi,\ \mu=2)$ and Lorentzian squared $(b=2/\pi,\ \mu=4)$] pulses, one finds that $z_c^0\sim(br)^{1/\mu}\exp[i\pi/(2\mu)]$ and

$$z_a\sim\beta+i\alpha(br)^{1/\mu}\left(\frac{1}{\mu-1}+\sum_{n=1}^{\infty}\frac{(2n-1)!!}{2^n n!(2n-1)(2\mu n-\mu+1)}\right)\exp\left(\frac{i\pi}{2\mu}\right). \quad (36)$$

On the other hand, for pulse shapes that vary as $b\exp[-at^\mu]$ $(\mu>1)$ when $|t|\gg 1$ [as do the hyperbolic secant $(b=1,\ a=\pi/2,\ \mu=1)$, hyperbolic secant squared $(b=\pi,\ a=\pi/2,\ \mu=1)$ and Gaussian $(b=1/\sqrt{\pi},\ a=1,\ \mu=2)$] pulses, one finds that $z_c^0\sim[\ln(rb)/a]^{1/\mu}$ and

$$z_a\sim\beta+i\frac{\pi\alpha}{2a\mu}\frac{1}{[\ln(rb)/a]^{1-1/\mu}}. \quad (37)$$

The corrections to Eqs. (36) and (37) depend on the specific form of the pulse shape.

For the specific pulse shapes considered in this paper one finds

$$a_2(\infty,hs)\sim-2ie^{-\alpha}\sin\beta, \quad (38a)$$

$$a_2(\infty,lz)\sim-2ie^{-1.20(\alpha\beta/\pi)^{1/2}}\sin[\beta-1.20(\alpha\beta/\pi)^{1/2}], \quad (38b)$$

$$a_2(\infty,hs^2)\sim-2ie^{-\alpha/2}\sin\beta, \quad (38c)$$

$$a_2(\infty,lz^2)\sim-2ie^{-0.416\alpha[4\beta/\pi\alpha]^{1/4}}\sin[\beta-0.172\alpha(4\beta/\pi\alpha)^{1/4}], \quad (38d)$$

$$a_2(\infty,gs)\sim-2ie^{-\alpha\pi/4\ln\sqrt{2\beta/\sqrt{\pi}\alpha}}\sin\beta. \quad (38e)$$

The corrections to the arguments of the exponentials and sine functions are of order $\alpha/\sqrt{r}$ for the Lorentzian pulse, α/r for the hyperbolic secant squared pulse, $\alpha/r^{1/4}$ for the Lorentzian squared pulse, and $\alpha/(\ln r)^{3/2}$ for the Gaussian pulse. The error correction for the Gaussian actually must be of order unity if the zeros z_c are to be well separated in the first quadrant of the complex plane. Thus the asymptotic form for the Gaussian cannot be considered to be totally reliable. Clearly, as the field strength approaches infinity, the magnitude of $a_2(\infty,gs)$ cannot exceed unity, whereas the asymptotic result (38e) increases (albeit very slowly) without limit.

The asymptotic forms (38) correctly map the large-β dependence of the transition probabilities shown in Fig. 1 and are in agreement with the conclusions reached in the qualitative discussion of the Massey parameter given in Sec. III. In fact, by comparing Eqs. (15) and (38), one finds that all the asymptotic results in the limit of large β are given by the relationship

$$a_2(\infty)\sim -2ie^{-K\mathrm{Im}\mathfrak{M}(z_c^0)}\sin[\beta-K\mathrm{Re}\mathfrak{M}(z_c^0)], \qquad (39)$$

where

$$\mathfrak{M}(z_c^0)\sim -\alpha/(r\ df/dz_c^0), \qquad (40)$$

is proportional to the minimum value of the Massey parameter when $r\gg 1$ and K is a constant of order unity that varies with pulse shape. We conjecture that Eq. (39) is a universal equation for the transition amplitude, even though it has been strictly proved only for pulse shapes that vary as $bt^{-\mu}$ $(\mu>1)$ or $b\exp[-at^{\mu}]$ $(\mu>1)$ when $|t|\gg 1$.

2. Series solution in r

When $r\ll 1$, one can expand Eq. (32) as

$$z_a\sim\alpha\int_0^{z_c^0}dz\left(1+\sum_{n=1}^{\infty}\frac{(-1)^{n+1}(2n-1)!![rf(z)]^{2n}}{2^n n!(2n-1)}\right). \qquad (41)$$

Using the values $z_c^0\sim\alpha[i+r/\pi+O(r^2)]$ for the hyperbolic secant pulse,

$$z_c^0\sim\alpha\left[i\left(1+\frac{r^2}{8\pi^2}\right)+\frac{r}{2\pi}+O(r^3)\right]$$

for the Lorentzian pulse,

$$z_c^0\sim\alpha\left[i-i\sqrt{\frac{r}{2\pi}}\left(1-\frac{\pi r}{24}\right)+\sqrt{\frac{r}{2\pi}}\left(1+\frac{\pi r}{24}\right)+O(r^{5/2})\right]$$

for the hyperbolic secant squared pulse,

$$z_c^0\sim\alpha\left[i-i\frac{1}{2}\sqrt{\frac{r}{\pi}}\left(1-\frac{r}{4\pi}\right)+\frac{1}{2}\sqrt{\frac{r}{\pi}}\left(1+\frac{r}{4\pi}\right)+\frac{r}{4\pi}+O(r^{5/2})\right]$$

for the Lorentzian squared pulse, and

$$z_c^0\sim\alpha\left[i|\ln(r/\sqrt{\pi})|^{1/2}+\frac{\pi}{4|\ln(r/\sqrt{\pi})|^{1/2}}+O(|\ln r|^{-3/2})\right]$$

for the Gaussian pulse, it follows from Eqs. (41) and (14) that for the specific pulse shapes considered in this paper

$$a_2(\infty,hs)\sim -2ie^{-\alpha}\sin\beta, \qquad (42a)$$

$$a_2(\infty,lz)\sim -2i\exp\left[-\alpha\left\{1+\left(\frac{6\ln 2-1-2\ln\left(\frac{2\beta}{\pi\alpha}\right)}{16}\right)\times\left(\frac{2\beta}{\pi\alpha}\right)^2\right\}\right]\sin\left[\frac{\beta}{2}\left\{1+\frac{1}{4}\left(\frac{2\beta}{\pi\alpha}\right)\right\}\right], \qquad (42b)$$

$$a_2(\infty,hs^2)\sim -2i\exp\left[-\alpha+\sqrt{\frac{2\alpha\beta}{\pi}}\left\{0.847-0.309\left(\frac{\pi\beta}{2\alpha}\right)\right\}\right]\sin\left[\sqrt{\frac{2\alpha\beta}{\pi}}\left\{0.847-0.309\left(\frac{\pi\beta}{2\alpha}\right)\right\}\right], \qquad (42c)$$

$$a_2(\infty,lz^2)\sim -2i\exp\left[-\alpha+\sqrt{\frac{\alpha\beta}{\pi}}\left\{0.847-0.232\left(\frac{4\beta}{\pi\alpha}\right)\right\}+\alpha\left(\frac{4\beta}{\pi\alpha}\right)^2\left\{0.0586-0.0391\ln\left(\frac{4\beta}{\pi\alpha}\right)\right\}\right]\times\sin\left[\sqrt{\frac{\alpha\beta}{\pi}}\left\{0.847+0.232\left(\frac{4\beta}{\pi\alpha}\right)\right\}+0.196\left(\frac{4\beta}{\pi\alpha}\right)\alpha+0.0614\left(\frac{4\beta}{\pi\alpha}\right)^2\alpha\right], \qquad (42d)$$

$$a_2(\infty,gs)\sim -2i\exp\left[-\alpha\left|\ln\left(\frac{2\beta}{\sqrt{\pi}\alpha}\right)\right|^{1/2}\times\left(1-\frac{0.153}{\left|\ln\left(\frac{2\beta}{\sqrt{\pi}\alpha}\right)\right|}\right)\right]\sin\left[\frac{\alpha\pi}{4\left|\ln\left(\frac{2\beta}{\sqrt{\pi}\alpha}\right)\right|^{1/2}}\right]. \qquad (42e)$$

The corrections to the arguments of the exponentials and sine functions are of order αr^3 for the Lorentzian pulse, $O(\alpha r^{5/2})$ for the hyperbolic secant squared pulse, $O(\alpha r^{5/2})$ for the Lorentzian squared pulse, and $O(\alpha|\ln r|^{-3/2})$ for the Gaussian pulse.

Several features are present in these results that one may consider to be puzzling. Equation (42a) for the hyperbolic secant pulse is the correct asymptotic result, despite the fact that the zeros in the first and second quadrants at $z_c\sim i\pm r$ coalesce as $r\sim 0$. For the Lorentzian pulse whose zeros $z_c\sim i\pm r/2$ also coalesce as $r\sim 0$, the term of order β, $a_2(\infty,lz)\sim -2i\beta e^{-\alpha}$, agrees with the perturbation theory result (20), but higher-order terms do not even have the same *functional* form. For both the hyperbolic secant squared and Lorentzian squared pulses, the lowest-order term in β does not agree with the perturbation theory result. For example, $a_2(\infty,hs^2)\sim -2ie^{-\alpha}(0.847\sqrt{2\alpha\beta/\pi})$, whereas the perturbation theory result (22) is $a_2(\infty,hs^2)\sim -2i\alpha\beta e^{-\alpha}$. This result is not surprising since, for the hyperbolic secant squared and Lorentzian squared pulses, there are *two* dominant zeros in the first quadrants that coalesce as $r\sim 0$. For the hyperbolic secant squared pulse, they occur at $z_c(hs^2) \sim i(1\pm\sqrt{\alpha\beta/\pi})+\sqrt{\alpha\beta/\pi}$. If one includes the contributions from each of these zeros independently, the asymptotic result becomes

$$a_2(\infty,hs^2)\sim -4ie^{-\alpha}\sin\left[0.847\sqrt{\frac{2\alpha\beta}{\pi}}\right]\sinh\left[0.847\sqrt{\frac{2\alpha\beta}{\pi}}\right]$$
$$\sim -1.83i\alpha\beta e^{-\alpha},$$

which has the correct functional form but differs from the perturbation theory result by about 9%. The sin sinh behavior has been predicted previously by Robinson and Berman [12] and shows that the probability envelope increases with increasing β for $\beta \ll \alpha$ ($r\ll 1$). For the Gaussian pulse, the lead term of the asymptotic expansion

$$a_2(\infty,gs)\sim -2i\exp\left[-\alpha\left|\ln\left(\frac{2\beta}{\sqrt{\pi}\alpha}\right)\right|^{1/2}\right]$$
$$\times\left(\frac{\alpha\pi}{4\left|\ln\left(\frac{2\beta}{\sqrt{\pi}\alpha}\right)\right|^{1/2}}\right)$$

has no resemblance to the perturbation theory result $a_2(\infty,gs)\sim -i\beta e^{-\alpha^2/4}$. The Gaussian pulse has an *infinite* number of zeros $z_c\sim\alpha i[|\ln(r/\sqrt{\pi})|-(2n+1)i\pi/2]^{1/2}$ $\{n,-\infty,\infty\}$, which coalesce as $r\sim 0$. It remains a challenge to modify the asymptotic results [3,17,18] so that they correctly reproduce the perturbation theory results. Moreover, it would be interesting to know why the asymptotic result for the hyperbolic secant pulse is essentially exact.

3. Numerical results

The transition probability $P_2=4e^{-2y_a^0}\sin^2 x_a^0$ for the various pulse shapes is shown as the dashed curves in Fig. 1, with x_a^0 and y_a^0 obtained by numerically integrating Eq. (32). Aside from significant relative differences in the solutions near the minima, which are not apparent in Fig. 1, the asymptotic and exact solutions are in very good agreement over the entire range of β shown in the graphs. To get some idea of the errors involved, we have plotted the ratio of the values of the envelopes of the asymptotic and exact solutions for $\alpha=5$ and several values of β in Fig. 2(a) and at fixed β for several values of α in Fig. 2(b). In general, the accuracy of the solution increases with increasing α or β as one might expect, but the rate of improvement depends on the specific pulse shape. The values of α and β in Fig. 2 are chosen to ensure that the zeros z_c in the first quadrant for the hyperbolic secant squared, Lorentzian squared, and Gaussian pulses are well separated, in accordance with one of the validity criteria for the asymptotic solution. For the hyperbolic secant pulse, the asymptotic and exact solutions differ by a negligible amount for $\alpha>5$; consequently, data for the hyperbolic secant pulse are omitted from Fig. 2. For the Lorentzian pulse, agreement between the exact and asymptotic solutions is good over the entire range of β, differing at most

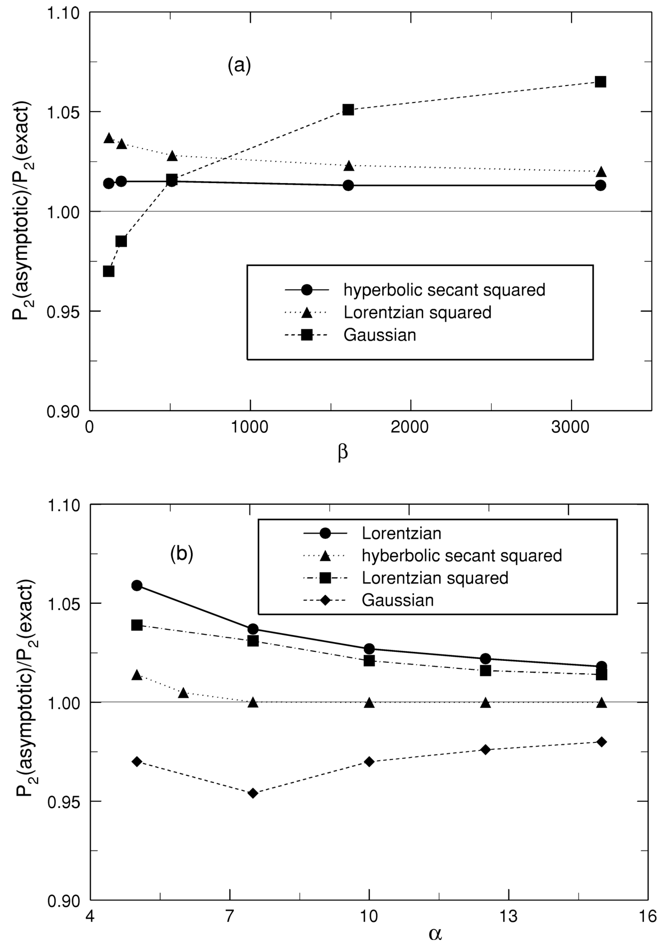

FIG. 2. Ratio of the envelope of the asymptotic solution of the differential equations given by Eqs. (31) and (32) for P_2 to the exact numerical results (a) as a function of β for $\alpha=5$ and (b) as a function of α for $\beta=2.8$ (Lorentzian pulse) and $\beta=75\pi/2$ (hyperbolic secant squared, Lorentzian squared, and Gaussian pulses).

by of order 6%. Values of $\beta>20$ have not been considered for the Lorentzian pulse owing to the long integration times required for the exact solution, but we expect the ratio of asymptotic to exact results to diminish with increasing β. For the hyperbolic secant squared and Lorentzian squared pulses, the asymptotic solution can be *orders of magnitude* larger than the exact solution for $\sqrt{\alpha\beta}\ll 1$ owing to the coalescing of the two zeros z_c in the first quadrant. A marked improvement in the solution for $\sqrt{\alpha\beta}\ll 1$ can be achieved if the second saddle point is properly incorporated into the problem. On the other hand, for $\sqrt{\alpha\beta}>1$, the asymptotic and exact solutions are in excellent agreement. The relative accuracy for the hyperbolic secant squared pulse is better than that for the Lorentzian squared pulse. For the Gaussian pulse, the asymptotic solution can again be orders of magnitude greater than the exact solution if $\alpha\sqrt{|\ln(\beta/\alpha|}\gg 1$, owing to the coalescing of an infinite number of zeros in the first quadrant. This condition occurs for *both* very small and very large-β. Some evidence for the large-β behavior is seen in Fig. 2(a), where the ratio of the asymptotic to exact solution grows with increasing β for $\beta\gtrsim 400$. Better agreement for large β can be obtained by using Crother's form (33) for the transition amplitude since it guarantees unitarity, but a proper treatment for very small and very large β requires one to include the contributions from the infinite number of zeros z_c in the complex plane.

V. SUMMARY

The nonadiabatic coupling of a two-level quantum system has been considered in detail. The specific model system was a two-level atom driven by an off-resonance radiation pulse, but the calculations apply to a large class of problems. It was shown that the dependence of the transition probability on coupling strength exhibits qualitatively different behavior for different pulse shapes. An explanation of this feature could be given in terms of the Massey parameter (ratio of the frequency separation of the semiclassical dressed states to the coupling strength in the semiclassical dressed-state basis). If the Massey parameter decreases with increasing coupling strength, the transition probability envelope increases with increasing field strength. In this limit the transition probability is no longer bound by a value based on the energy-time uncertainty principle. The uncertainty principle argument, in which the transition probability is exponentially small in the dimensionless detuning parameter α, is associated with first-order perturbation theory (Fourier transform of the pulse). Its extension to the nonlinear domain depends on the pulse shape and requires a pulse shape for which the Massey parameter increases or remains constant with increasing coupling strength β for $\beta \gg 1$.

Numerical and asymptotic solutions have been presented. For $\alpha \gg 1$, the asymptotic solution of the differential equations yields results that are in excellent agreement with the exact result, provided that β is limited to values (usually $\beta \gtrsim 1$) for which there are no coalescing zeros z_c in the first quadrant of the complex plane of the function $[\alpha^2 + 4\beta^2 f^2(z)]$. However, several anomalies in the asymptotic solutions have been noted. In particular, there does not appear to be a correct asymptotic evaluation of the integral (27) encountered in first-order perturbation theory in the dressed basis. Moreover, there does not appear to be any systematic way in which the asymptotic solutions of the differential equations approach the results of perturbation theory.

The results derived in this work could be tested using pulsed radiation fields interacting with atoms. Two types of experiments can be envisioned. In the first, an ultrafast radiation pulse ($\sim$100 fs) is incident on atoms in a vapor cell. The pulse duration must be much less than the excited state lifetime and pulse areas 2β greater than unity should be accessible, but the pulse strength should not be so large as to ionize the atom. With modest power density ($\sim 10^{10}-10^{12}$ W/cm^2), values of β on the order of 20 can be achieved. The second type of experiment is one in which a beam of atoms passes through a cw laser field. If one chooses an atom such as ytterbium for which the resonance transition has a lifetime of order 1 μs, large pulse areas can be realized by using relatively long interaction times [19]. The advantage of this method is that the pulse shape in the atom's rest frame will be Gaussian if the spatial profile of the laser field is Gaussian.

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APPENDIX

Several details of the calculations are presented below.

1. Perturbation series solution

Equations (1) can be solved iteratively. To first order in β, one finds

$$a_2^{(1)} = -i\beta \int_{-\infty}^{\infty} dt\, e^{-i\alpha t} f(t). \tag{A1}$$

The third-order contribution is

$$a_2^{(3)} = (-i\beta)^3 \int_{-\infty}^{\infty} dt e^{-i\alpha t} f(t) \int_{-\infty}^{t} dt' e^{i\alpha t'} f(t') \times \int_{-\infty}^{t'} dt'' e^{-i\alpha t''} f(t''). \tag{A2}$$

To obtain an asymptotic expansion for large α, we integrate by parts n times to arrive at

$$a_2^{(3)} \sim (-i\beta)^3 \int_{-\infty}^{\infty} dt\, e^{-i\alpha t} f(t) \int_{-\infty}^{t} dt' e^{i\alpha t'} f(t') \times \sum_{n=1}^{\infty} (-i\alpha)^{-n} (-1)^{n+1} f^{(n-1)}(t')\, e^{-i\alpha t'}. \tag{A3}$$

It is worth noting two things at this point. First, *all* terms in the sum must be retained. The higher-order derivatives $f^{(n-1)}$ lead to contributions that are higher order in α, compensating for the $(-i\alpha)^{-n}$ dependence. Second, the integration by parts technique does not work for the first-order contribution [Eq. (A1)] since all terms in the asymptotic series vanish at $t=\infty$. By interchanging the order of integration, one finds

$$a_2^{(3)} \sim (-i\beta)^3 \int_{-\infty}^{\infty} dt\, f(t) \sum_{n=1}^{\infty} (-i\alpha)^{-n} (-1)^{n+1} f^{(n-1)}(t) \times \int_{t}^{\infty} dt\, e^{-i\alpha t} f(t). \tag{A4}$$

Again integrating by parts m times, one finally obtains

$$a_2^{(3)} \sim (-i\beta)^3 \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} (-1)^{n+m+1} (-i\alpha)^{-(n+m)} \times \int_{-\infty}^{\infty} dt\, f(t)\, f^{(n-1)}(t)\ \ f^{(m-1)}(t)\, e^{-i\alpha t}. \tag{A5}$$

The fifth-order contribution is

$$a_2^{(5)}=(-i\beta)^5\int_{-\infty}^{\infty}dt\ e^{-i\alpha t}\ f(t)\int_{-\infty}^{t}dt' e^{i\alpha t'}\ f(t')$$
$$\times\int_{-\infty}^{t'}dt'' e^{-i\alpha t''}\ f(t'')\int_{-\infty}^{t''}dt'''\ e^{i\alpha t'''}\ f(t''')$$
$$\times\int_{-\infty}^{t^{iv}}dt^{iv}\ e^{-i\alpha t^{iv}}\ f(t^{iv}). \tag{A6}$$

Using techniques similar to those used for the third-order contribution, one can obtain

$$a_2^{(5)}\sim(-i\beta)^5\sum_{n,m,p,q=1}^{\infty}(i\alpha)^{-(n+m+p+q)}\int_{-\infty}^{\infty}dt\ f(t)$$
$$\times f^{(n-1)}(t)e^{-i\alpha t}\ \frac{d^{m-1}}{dt^{m-1}}[f(t)\ f^{(p-1)}(t)\ \ f^{(q-1)}(t)]. \tag{A7}$$

It is now possible to carry out explicit evaluations of Eqs. (A1), (A5), and (A7) for the $f_j(t)$ $(j=1-5)$ considered in this work. In doing so, one obtains the leading terms in α^{-1} by approximating the derivatives as $f_1^{(n)}(t)\sim(1/2)(-\pi/2)^n n!\sinh^n(\pi t/2)\cosh^{-(n+1)}(\pi t/2)$, $f_2^{(n)}(t)\sim(1/\pi)(-2t)^n n!(1+t^2)^{-(n+1)}$, $f_3^{(n)}(t)\sim(\pi/4)(-\pi/2)^n(n+1)!\sinh^n(\pi t/2)\cosh^{-(n+2)}(\pi t/2)$, $f_4^{(n)}(t)\sim(2/\pi)(-2t)^n(n+1)!(1+t^2)^{-(n+2)}$, and $f_5^{(n)}(t)\sim(1/\sqrt{\pi})(-2t)^n\exp(-t^2)$. Using these expressions in Eqs. (A1), (A5), and (A7), one finds

$$a_2(\infty,hs)=-i\ \mathrm{sech}\ \alpha\biggl(\beta-\frac{\beta^3}{3!}[1+O(\alpha^{-2})]$$
$$+\frac{\beta^5}{5!}[1+O(\alpha^{-2})]-\cdots\biggr), \tag{A8a}$$

$$a_2(\infty,lz)=-2ie^{-\alpha}\biggl[\frac{\beta}{2}-\left(\frac{\beta}{2}\right)^3\left(\frac{1}{3!}\right)[1+O(\alpha^{-2})]$$
$$+\left(\frac{\beta}{2}\right)^5\left(\frac{1}{5!}\right)[1+O(\alpha^{-2})]-\cdots\biggr], \tag{A8b}$$

$$a_2(\infty,hs^2)=-i\pi\ \mathrm{csch}\ \alpha\biggl[\left(\frac{\alpha\beta}{\pi}\right)-\left(\frac{\alpha\beta}{\pi}\right)^3\left(\frac{10-\pi^2}{6}\right)$$
$$\times[1+O(\alpha^{-2})]+3.505\times10^{-5}\left(\frac{\alpha\beta}{\pi}\right)^5$$
$$\times[1+O(\alpha^{-2})]-\cdots\biggr]. \tag{A8c}$$

$$a_2(\infty,lz^2)=-2i\pi e^{-\alpha}\biggl[\left(\frac{\alpha\beta}{2\pi}\right)\left(1+\frac{1}{\alpha}\right)-\left(\frac{\alpha\beta}{2\pi}\right)^3\left(\frac{10-\pi^2}{6}\right)$$
$$\times[1+O(\alpha^{-2})]+3.505\times10^{-5}\left(\frac{\alpha\beta}{2\pi}\right)^5$$
$$\times[1+O(\alpha^{-2})]-\cdots\biggr], \tag{A8d}$$

$$a_2(\infty,gs)=-ie^{-\alpha^2/4}\biggl[\beta e^{-\alpha^2/4}-\left(\frac{9\beta^3e^{-\alpha^2/12}}{4\sqrt{3}\pi\alpha^2}\right)$$
$$\times[1+O(\alpha^{-2})]+\frac{625\beta^5e^{-\alpha^2/20}}{64\sqrt{5}\pi^2\alpha^4}[1+O(\alpha^{-2})]$$
$$-O(\beta^7\alpha^{-6}e^{-\alpha^2/28})\biggr]. \tag{A8e}$$

2. First-order perturbative solution in the dressed basis: Series solution in β

The first-order perturbative solution in the dressed basis (27) can also be expanded as a power series in β. The first-order contribution is

$$a_2^{(1,1)}(ad)=-(\beta/\alpha)\int_{-\infty}^{\infty}dt\ e^{-i\alpha t}df/dt$$
$$=-i\beta\int_{-\infty}^{\infty}dt\ e^{-i\alpha t}\ f(t), \tag{A9}$$

which is identical to the first term in the perturbative expansion of the exact equations. The superscript $(1,n)$ indicates that the result is the β^n term in the expansion of the first-order perturbative solution of the dressed-state equations (26) (recall that the first-order perturbative solution in the dressed-state basis contains all powers of β). The third-order contribution is

$$a_2^{(1,3)}(ad)=(\beta^3/\alpha^3)\int_{-\infty}^{\infty}dt\ e^{-i\alpha t}\frac{df}{dt}$$
$$\times\left(4f^2+2i\alpha\int_0^t dt'\ [f(t')]^2\right) \tag{A10}$$

and the fifth-order contribution is

$$a_2^{(1,5)}(ad)=-(\beta^5/\alpha^5)\int_{-\infty}^{\infty}dt\ e^{-i\alpha t}\frac{df}{dt}$$
$$\times\biggl[16f^4+8i\alpha f^2\int_0^t dt'\ [f(t')]^2$$
$$+8i\alpha\int_0^t dt'\ [f(t')]^4$$
$$-2\alpha^2\left(\int_0^t dt'\ [f(t')]^2\right)^2\biggr]. \tag{A11}$$

For the various pulse shapes functions $f_j(t)$ $(j=1-5)$, we have evaluated the β^3 terms to all orders in α^{-1} and the β^5 terms to lowest nonvanishing order in α^{-1}.

Most of the integrals are tabulated or can be done by contour integration. However, we encountered a few integrals that do not appear in standard integral tables. For the Lorentzian and Lorentzian-squared pulses, one must evaluate integrals of the form $\int_{-\infty}^{\infty}dt\ e^{-i\alpha t}\tan^{-1}t\ /(1+t^2)^n$ with n

n=1,2. These integrals can be evaluated by defining $I(b) = \int_{-\infty}^{\infty} dt\ e^{-i\alpha t}\tan^{-1}(bt)\ /(1+t^2)^n$, differentiating with respect to b (at which point the integral over time can be carried out), integrating the result over b from 0 to $1-\epsilon$, and taking the limit $\epsilon \sim 0$. In this manner one finds

$$\int_{-\infty}^{\infty} dt\ e^{-i\alpha t}\tan^{-1}t\ /(1+t^2) = -i(\pi/2)e^{-\alpha}[M(\alpha) + N(\alpha)], \tag{A12a}$$

$$\int_{-\infty}^{\infty} dt\ e^{-i\alpha t}\tan^{-1}t\ /(1+t^2)^2 = -i(\pi/4)e^{-\alpha}[-\alpha + (1+\alpha)M(\alpha) + (1-\alpha)N(\alpha)], \tag{A12b}$$

where

$$M(\alpha) = C + \ln 2 + \ln\alpha,$$
$$N(\alpha) = \left(\frac{1}{2\alpha} - \frac{1}{4\alpha^2} + \frac{2}{8\alpha^3} - \cdots\right), \tag{A13}$$

$C \simeq 0.577$ is Euler's constant, and $N(\alpha)$ is the asymptotic series $\Sigma_{k=1}^{\infty}(-1)^{k+1}(k-1)!/(2\alpha)^k$. For the Gaussian pulse, one encounters integrals of the form $\int_{-\infty}^{\infty} dt\ te^{-i\alpha t}e^{-t^2}\int_0^t dt' e^{-2t'^2}$ and $\int_{-\infty}^{\infty} dt\ te^{-i\alpha t}e^{-t^2}(\int_0^t e^{-2t'^2}dt')^2$. The first of these can be calculated by interchanging the order of integration and using integration by parts. The second can be calculated by an n-fold integration by parts in which, at each step, one replaces $d/dt[t^n e^{-t^2}(\int_0^t dt'\ e^{-2t'^2})^2]$ by $-2t^{n+1}e^{-t^2}(\int_0^t dt'\ e^{-2t'^2})^2 + t^n e^{-3t^2}\int_0^t dt' e^{-2t'^2}$ to get the leading terms in α^{-1}. In this manner one finds

$$\int_{-\infty}^{\infty} dt\ te^{-i\alpha t}e^{-t^2}\int_0^t dt' e^{-2t'^2} = \left(\frac{\sqrt{\pi}}{4\sqrt{3}}\right)e^{-\alpha^2/12}[1+S(\alpha)], \tag{A14a}$$

$$\int_{-\infty}^{\infty} dt\ te^{-i\alpha t}e^{-t^2}\left(\int_0^t e^{-2t'^2}dt'\right)^2 = \left(\frac{i\sqrt{5\pi}}{8\alpha}\right)e^{-\alpha^2/20}[1+O(\alpha^{-2})], \tag{A14b}$$

where

$$S(\alpha) = \sqrt{6}e^{\alpha^2/12}\int_{-\infty}^{\infty} dt\ te^{-i\alpha t}e^{-t^2}\mathrm{erf}(\sqrt{2}t) + 1 \tag{A15}$$

can be developed as an asymptotic series in α^{-2} whose lead term is approximately equal to $-9.4/\alpha^2$. With these results, one can carry out the integrations in Eqs. (A9)–(A11) to obtain

$$a_2^{(1)}(\infty, hs; ad) = -i\,\mathrm{sech}\alpha\left[\beta - \frac{\beta^3}{3!}\left(\frac{10}{\pi^2}\right)\left(1 - \frac{\pi^2}{20\alpha^2}\right) + \frac{\beta^5}{5!}\left(\frac{10}{\pi^2}\right)^2\left(\frac{298}{300}\right)[1+O(\alpha^{-2})] - \cdots\right], \tag{A16a}$$

$$a_2^{(1)}(\infty, lz; ad) = -2ie^{-\alpha}\left\{\frac{\beta}{2} - \left(\frac{\beta}{2}\right)^3\left(\frac{1}{3!}\right)\left(\frac{10}{\pi^2}\right)\left[1 - \frac{3}{5\alpha} - \frac{3}{5\alpha^2} + M(\alpha) + N(\alpha)\right] + \left(\frac{\beta}{2}\right)^5\left(\frac{1}{5!}\right) \times \left(\frac{10}{\pi^2}\right)^2\left(\frac{298}{300}\right)[1+O(\alpha^{-1})] - \ \ldots\right\}, \tag{A16b}$$

$$a_2^{(1)}(\infty, hs^2; ad) = -i\pi\,\mathrm{csch}\alpha\left[\left(\frac{\alpha\beta}{\pi}\right) - \left(\frac{\alpha\beta}{\pi}\right)^3\left(\frac{16}{6!}\right) \times \left(1 + \frac{15\pi^2}{2\alpha^2} - \frac{\pi^4}{\alpha^4}\right) + \left(\frac{16}{9!}\right)\left(\frac{516}{630}\right)\left(\frac{\alpha\beta}{\pi}\right)^5 \times [1+O(\alpha^{-2})] - \cdots\right], \tag{A16c}$$

$$a_2^{(1)}(\infty, lz^2; ad) = -2i\pi e^{-\alpha}\left[\left(\frac{\alpha\beta}{2\pi}\right)\left(1 + \frac{1}{\alpha}\right) - \left(\frac{\alpha\beta}{2\pi}\right)^3\left(\frac{16}{6!}\right) - \left(\frac{\alpha\beta}{2\pi}\right)^3\left(\frac{16}{6!}\right)\left(1 + \frac{520}{32\alpha} + \frac{4560}{32\alpha^2} + \frac{2040}{32\alpha^3} - \frac{7560}{32\alpha^4} - \frac{7560}{32\alpha^5} + \frac{7200}{32\alpha^4} \times [-\alpha + (1+\alpha)M \times (\alpha) + (1-\alpha)N(\alpha)]\right) + \left(\frac{2^4}{9!}\right)\left(\frac{516}{630}\right) \times \left(\frac{\alpha\beta}{2\pi}\right)^5 [1+O(\alpha^{-1})] - \cdots\right], \tag{A16d}$$

$$a_2^{(1)}(\infty, gs; ad) = -i\left[\beta e^{-\alpha^2/4} - \left(\frac{7\beta^3 e^{-\alpha^2/12}}{3\sqrt{3}\pi\alpha^2}\right) \times \left[1 - \frac{3}{7}S(\alpha)\right] + \left(\frac{51}{5}\right)\frac{\beta^5 e^{-\alpha^2/20}}{\sqrt{5}\pi^2\alpha^4} \times [1+O(\alpha^{-2})] - O(\beta^7\alpha^{-6}e^{-\alpha^2/28})\right]. \tag{A16e}$$

[1] For a detailed discussion of this problem, with many references to earlier work, see E. E. Nikitin and S. Ya. Umanskii, *Theory of Slow Atomic Collisions* (Springer-Verlag, Berlin, 1984), especially Chaps. 6–8.

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[9] Since we start in state 1 at $t=-\infty$, the transition probability P_2 is independent of the phase associated with the state 1 probability amplitude at $t=-\infty$.

[10] N. F. Mott and H. S. W. Massey, *The Theory of Atomic Collisions* (Oxford University Press, Oxford, 1965), Chaps. 19 and 21.

[11] Although not shown on the graph, the calculation was extended to $\beta=4000$, for which $P_2(\max)\simeq 0.2$

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[13] For the Gaussian pulse, the Massey parameter can actually become less than unity for extremely large values of β, such that $(1/2^{3/2}\alpha)\sqrt{\ln(2\beta/\sqrt{\pi})\alpha}>1$ (for $\alpha=5$, this condition requires $\beta>3.2\times 10^{87}$). In this limit, transitions are no longer nonadiabatic in nature and the arguments used to predict the dependence of the transition probability on the Massey parameter are no longer valid. In this regime, we expect the maximum transition probability to approach unity.

[14] It is possible to estimate the validity criteria for Eqs. (21)–(23) by comparing these equations with Eqs. (42b)–(42d). One sees that Eq. (42b) is consistent with Eq. (21) provided $\beta^2/\alpha\ll 1$. Equation (42c), modified to include the contribution from the second saddle point in the first quadrant of the complex plane, will have the same functional form as Eq. (22) provided $r\sqrt{\alpha\beta}\ll 1$. Equation (42d), modified to include the contribution from the second saddle point in the first quadrant of the complex plane, will have the same functional form as Eq. (23) provided $r<1$, $r\beta\ll 1$, and $r\sqrt{\alpha\beta}\ll 1$.

[15] The asymptotic results are given to within an overall phase factor that is chosen to match the perturbative solutions.

[16] For the hyperbolic and hyperbolic squared pulses, there are an infinite number of zeros in the first quadrant; however, it is sufficient to consider only those zeros that map onto $z_c\sim i$ as $\beta\sim 0$ since the other zeros contribute a negligibly small amount when $\alpha\gg 1$.

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